

Atty. Docket No. KOV-004

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF:

:

Klaus KUNZE et al.

: GROUP ART UNIT: 2822

APPLICATION NO: 10/616,147

:

FILED: July 8, 2003

: EXAMINER: TRINH, Michael Manh

FOR: COMPOSITIONS AND METHODS FOR FORMING A SEMICONDUCTING  
AND/OR SILICON-CONTAINING FILM, AND STRUCTURES FORMED  
THEREFROM

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By: /Judy Ryan/  
Judy Ryan

DECLARATION UNDER 37 C.F.R. 1.132

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SIR:

Now comes Fabio ZÜRCHER, who declares and states that:

1. I have been employed by Kovio, Inc., as the Manager of Ink Development since October 16, 2001. Prior to that time, I was employed by Department of Chemistry at the University of California, Berkeley, as a postdoctoral fellow, where my duties included research in the area(s) of synthesis of silicon nanoparticles.

2. I received a Bachelor's degree in Chemistry from the ETH Zurich (Swiss Federal Institute of Technology), Switzerland, in chemistry, and a Ph.D. in chemistry from the ETH Zurich (Swiss Federal Institute of Technology), Switzerland.

3. I have read the above-identified application and am familiar with the subject matter disclosed and claimed therein. I have reviewed Shiho et al. (U.S. Pat. App. Pub. No. 2003/0045632 [hereinafter, "Shiho"]); Jacobson et al. (U.S. Pat. No. 6,294,401 [hereinafter, "Jacobson '401']"); Piwczyn (U.S. Pat. No. 4,022,928 [hereinafter, "Piwczyn"]); Beppu et al. (U.S. Pat. No. 5,866,471 [hereinafter, "Beppu"]); Tani et al. (U.S. Pat. No. 5,254,439 [hereinafter, "Tani"]); Kim et al. (U.S. Pat. No. 6,355,198 [hereinafter, "Kim"]); and Korgel (U.S. Pat. App. Pub. No. 2003/0034486 [hereinafter, "Korgel"]). I am also familiar with silane chemistry, silicon and metal nanoparticles, and techniques for making thin films using the same.

4. I understand that one independent claim of the above-identified application is directed to a method of making a patterned semiconductor film, comprising the steps of:

- a) printing a solution comprising silicon-containing semiconductor nanoparticles having a passivation layer covalently bound thereto selected from the group consisting of an alcohol, an alcoholate, a thiol, a thiolate, an alkyl group, and an aralkyl group, a first cyclic Group IVA compound of the formula (1):



where n is from 3 to 8, each of the n instances of x is independently 1 or 2, and each A in the formula is independently Si or Ge, and a solvent in a pattern on a substrate; and

- b) curing said printed pattern to form said patterned semiconductor film.

5. Shiho discloses silane compositions for preparing semiconductor thin films in solar cells (Abstract). Shiho further discloses several semiconductor thin films that may be formed from the silane compositions disclosed.

6. For example, Shiho discloses a series of four components A – D that are the essential components of the compositions disclosed, and may be combined in various ways to form compositions, as described below.

7. Component A is represented by the formula  $Si_nR_m$ , with R substituents being selected from hydrogen, alkyl, phenyl or halogens (see paragraph [0040] of Shiho). Component B is at least one silane compound selected from the group consisting of cyclopentasilane, cyclohexasilane and silylcyclopentasilane (see paragraph [0050] of Shiho). Component C is silicon particles (see paragraph [0060] of Shiho), which may have a particle diameter of 0.005 to 1,000  $\mu m$  (see paragraph [0061] of Shiho), and may be amorphous or crystalline (see paragraph [0063] of Shiho). Component D is at least one boron compound, arsenic compound, phosphorus compound, antimony compound or modified silane compound represented by the formula  $Si_aX_bY_c$  (where X is a hydrogen atom and/or halogen atom, Y is a boron atom or phosphorus atom, a is an integer of 3 or more, b is an integer of 1 or more and a or less, and c is an integer of a or more and  $(2a+b+2)$  or less, see paragraph [0076]).

8. Shiho is silent with respect to silicon-containing semiconductor nanoparticles having a passivation layer covalently bound thereto selected from the group consisting of an alcohol, an alcoholate, a thiol, a thiolate, an alkyl group, and an aralkyl group (see paragraph 4, above).

9. Silicon-containing semiconductor nanoparticles typically exhibit low solubility in conventional organic solvents. As a result, liquid-phase compositions comprising silicon-containing semiconductor nanoparticles often have insufficient mass loading of the silicon-containing semiconductor nanoparticles to make a semiconductor film therefrom.

10. Unpassivated silicon particles such as those disclosed by Shiho (see paragraph [0061] of Shiho; see also paragraph 7 above) generally exhibit very low solubility or substantial insolubility in certain organic solvents (e.g., hydrocarbons, arenes and ethers).

11. I conducted, supervised, witnessed or otherwise know from the ordinary course of business at Kovio a number of experiments performed at Kovio to test the physical and chemical properties of hydrogen-passivated silicon nanoparticles.

12. A technical report presented by my colleagues and I to Kovio, Inc. management during the ordinary course of business (entitled *Nanocrystalline Silicon Ink*; attached hereto as Exhibit A) describes certain physical properties of hydrogen-passivated silicon nanoparticles.

13. Compositions comprising the hydrogen-passivated silicon nanoparticles and certain organic solvents (e.g., butyl ether, xylene, decalin and others; see page 6 of Exhibit A) were found to exhibit very low mass loadings of hydrogen-passivated silicon nanoparticles (< 0.1 %; see page 5 of Exhibit A).

14. Hydrogen-passivated silicon nanoparticles were expected to exhibit sufficient solubility in certain organic solvents (e.g., hydrocarbons, arenes and ethers) to enable printing of an ink containing such hydrogen-passivated silicon nanoparticles. However, the low mass loadings of hydrogen-passivated silicon nanoparticles in compositions comprising the hydrogen-passivated silicon nanoparticles and certain organic solvents (as described in paragraph 13 above) indicate that hydrogen-passivated silicon nanoparticles have insufficient solubility in such solvents to enable printing of an ink containing such hydrogen-passivated silicon nanoparticles.

15. I also conducted, supervised, witnessed or otherwise know from the ordinary course of business at Kovio a number of experiments performed at Kovio to test the physical and chemical properties of compositions containing alkyl-passivated silicon nanoparticles.

16. A technical report presented by my colleagues and I to Kovio, Inc. management during the ordinary course of business (entitled *Surface Derivatization of Silicon Nanocrystals*; attached hereto as Exhibit B) describes the preparation of alkyl-passivated silicon nanoparticles (see page 3 of Exhibit B). The report also describes certain physical and chemical properties of alkyl-passivated silicon nanoparticles.

17. Dodecyl-passivated silicon nanoparticles were chosen as representative alkyl-passivated nanoparticles (see page 3 of Exhibit B). The observed physical and chemical properties of dodecyl-passivated silicon nanoparticles are expected to be representative of alkyl-terminated silicon nanoparticles in general.

18. Alkyl-passivated silicon nanoparticles were found to be very soluble in certain organic solvents (e.g., hydrocarbons and ethers; see page 2 of Exhibit B).

19. Another technical report presented by my colleagues and I to Kovio, Inc. management during the ordinary course of business (entitled *Silicon Film Formation From Nanocrystals*; attached hereto as Exhibit C) reports experimental results quantifying the solubility of alkyl-passivated silicon nanoparticles in certain solvents (e.g., hydrocarbons and ethers).

20. Alkyl-passivated nanoparticles exhibited a solubility of up to 5% in xylene, and greater than 5% in butyl ether (see page 2 of Exhibit C).

21. Alkyl-passivated silicon nanoparticles exhibited on the order of two orders of magnitude greater solubility (e.g., 5 % or greater) relative to hydrogen-passivated silicon nanoparticles (e.g., less than 0.1 %).

22. The data, results and observations described in paragraphs 13-14, 18, 20 and 21 above and the disclosure of Shiho establish the superior solubility of alkyl-passivated silicon nanoparticles relative to hydrogen-passivated silicon nanoparticles (e.g., as described in paragraphs 11-14, above) or unpassivated silicon particles (e.g., those disclosed by Shiho; see paragraph 7 above).

23. Silicon-containing semiconductor nanoparticles having a passivation layer covalently bound thereto selected from the group consisting of an alcohol, an alcoholate, a thiol, a thiolate, and an aralkyl group (see paragraph 4 above) are expected to exhibit solubility analogous to that exhibited by alkyl-passivated silicon nanoparticles (see paragraph 20 above).

24. Shiho is silent with respect to silicon-containing semiconductor nanoparticles having a passivation layer covalently bound thereto selected from the group consisting of an alcohol, an alcoholate, a thiol, a thiolate, an alkyl group, and an aralkyl group (see paragraph 4, above). Therefore, Shiho cannot recognize the superior solubility of silicon-containing semiconductor nanoparticles having a passivation layer covalently bound thereto selected from the group consisting of an alcohol, an alcoholate, a thiol, a thiolate, an alkyl group, and an aralkyl group (see paragraph 4 above) relative to unpassivated silicon particles and hydrogen-passivated silicon nanoparticles (see paragraphs 22 and 23 above).

25. Jacobson '401 discloses a method for making electronic, chemical, and mechanical devices by deposition and patterning nanoparticles through printing technology (Abstract). Representative classes of nanoparticles include insulators (e.g., silicon dioxide); semiconductors (e.g., silicon or cadmium selenide); and conductors (e.g., silver) (col. 3, ll. 39-42).

26. Jacobson '401 further discloses that nanoparticles may be passivated at the surface by an organic capping group which surrounds the typically inorganic particle core as a shell. In such circumstances, it is the capping group that largely determines the solubility of the particles, and thus what solvents are appropriate for carrying the particles (col. 4, ll. 47-52). However, Jacobson '401 fails to disclose any solvents appropriate for solubilizing such inorganic particles having an organic capping group.

27. Jacobson '401 does not explicitly disclose silicon-containing semiconductor nanoparticles having a passivation layer covalently bound thereto selected from the group consisting of an alcohol, an alcoholate, a thiol, a thiolate, an alkyl group, and an aralkyl group (see paragraph 4 above). Furthermore, Jacobson '401 is silent with respect to cyclic Group IVA compounds.

28. The solubility of silicon-containing semiconductor nanoparticles is important for enabling printing of a liquid-phase composition comprising the silicon-containing semiconductor

nanoparticles in methods for making patterned semiconductor films. For example, insufficient mass loading of silicon-containing semiconductor nanoparticles in a liquid-phase ink composition comprising the silicon-containing semiconductor nanoparticles may result in a failure to produce a patterned semiconductor film therefrom due to beading and/or uneven spreading of the printed composition. Such beading and/or uneven spreading of the ink composition can also result in unacceptable film morphology and/or pattern uniformity and reproducibility.

29. In general, for many printed electronic devices a semiconductor precursor ink is printed on a non-wetting or non-absorbing. Films formed from such inks on such substrates will exhibit unacceptable pattern variations without a mechanism for controlling pattern uniformity. One example of a mechanism that controls pattern uniformity is including a material in the ink (e.g., nanoparticles) that precipitates from the ink composition before the printed pattern can bead or spread to a significant extent/degree.

30. As the solvent evaporates from a printed ink containing semiconductor nanoparticles, the semiconductor nanoparticles begin to precipitate out of the ink, and may prevent the printed pattern from beading or spreading too unevenly. The greater the mass loading of nanoparticles in the composition, the greater the probability that the nanoparticles will precipitate before significant variations in a printed pattern (e.g., as a result of beading or spreading) can occur.

31. Because of their very low solubility or substantial insolubility in solvents typically used for printable ink compositions, unpassivated silicon nanoparticles (see paragraph 9 above) or hydrogen-passivated silicon nanoparticles (see paragraph 14 above) by themselves exhibit insufficient mass loading in such compositions for use in methods of making patterned semiconductor films.

32. The superior solubility of alkyl-passivated silicon nanoparticles (see paragraph 23 above) enables higher mass loading of silicon-containing semiconductor nanoparticles, and

accordingly enables the formation of patterned semiconductor films via printing of compositions comprising alkyl-passivated silicon nanoparticles.

33. Neither Shiho nor Jacobson '401 disclose, suggest, or recognize that solubility (and corresponding mass loading) of semiconductor nanoparticles is a result-critical parameter in making patterned semiconductor films by printing compositions comprising semiconductor particles.

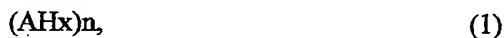
34. Neither Shiho nor Jacobson '401 disclose, suggest, or recognize that silicon-containing semiconductor nanoparticles having a passivation layer covalently bound thereto selected from the group consisting of an alcohol, an alcoholate, a thiol, a thiolate, an alkyl group, and an aralkyl group (see paragraphs 4, 22 and 23 above) exhibit superior solubility on the order of two orders of magnitude relative to hydrogen-passivated silicon nanoparticles (see paragraph 22 above). Furthermore, neither Shiho nor Jacobson recognize such superior solubility enables printing of compositions comprising alkyl-passivated silicon nanoparticles in methods for making patterned semiconductor films from such compositions.

35. As a result, no combination of Shiho and Jacobson '401 suggests a method of making a patterned semiconductor film including printing a solution comprising silicon-containing semiconductor nanoparticles having a passivation layer covalently bound thereto selected from the group consisting of an alcohol, an alcoholate, a thiol, a thiolate, an alkyl group, and an aralkyl group, a cyclic Group IVA compound and a solvent in a pattern on a substrate (see paragraph 4 above).



36. I understand that a second independent claim of the above-identified application is directed to a method of making a patterned semiconductor film, comprising the steps of:

- a) printing a solution comprising passivated semiconductor nanoparticles, a first cyclic Group IVA compound of the formula (1):



where n is from 3 to 8, each of the n instances of x is independently 1 or 2, and each A in the formula is independently Si or Ge, and a cycloalkane solvent in a pattern on a substrate; and

- b) curing said printed pattern to form said patterned semiconductor film.

37. One problem associated with making semiconductor films from compositions comprising cyclic Group IVA compounds, including those containing 5 or more silicon atoms in the cyclic ring (hereinafter, "Si<sub>5+</sub> cyclosilanes") is the stability of Si<sub>5+</sub> cyclosilanes in those solutions. The stability of compositions containing Si<sub>5+</sub> cyclosilanes is important for enabling the printability of such compositions in making semiconductor films from such compositions (which may also include passivated silicon-containing semiconductor nanoparticles).

38. I conducted, supervised, witnessed or otherwise know from the ordinary course of business at Kovio a number of experiments performed at Kovio to test the stability of Si<sub>5+</sub> cyclosilanes in various solvents.

39. A technical report presented by my colleagues and I to Kovio, Inc. management during the ordinary course of business (entitled *Polysilane Ink Formulation*, attached hereto as Exhibit D) reports the results of tests performed to determine the stability of Si<sub>5+</sub> cyclosilanes in certain solvents.

40. Si<sub>5+</sub> cyclosilanes exhibited improved stability in some cycloalkanes (cyclohexane, cycloheptane and cis-decalin) relative to arenes (benzene, toluene, and xylene) and other solvents such as linear alkanes, alcohols, ketones, alkenes, ethers, and fluorinated solvents (see page 3 of Exhibit D).

41. The improved stability of  $\text{Si}_{5+}$  cyclosilanes in some cycloalkane solvents relative to arene solvents (as described in paragraph 40 above) was unexpected in view of the published literature reporting mixtures of  $\text{Si}_{5+}$  cyclosilane(s) in other solvents, typically an arene (namely, toluene).

42. I also conducted, supervised, witnessed or otherwise know from the ordinary course of business at Kovio a number of experiments performed at Kovio to test the printability of  $\text{Si}_{5+}$  cyclosilanes in various solvents.

43. A technical report presented by my colleagues and I to Kovio, Inc. management during the ordinary course of business (entitled *Silane Ink Formulation: Solvent Selection and Controlled Polymerization*; attached hereto as Exhibit E) reports the results of tests performed to determine the printability of  $\text{Si}_{5+}$  cyclosilanes in various solvents, including results of controlled ultraviolet light-initiated polymerization of spincoated  $\text{Si}_{5+}$  cyclosilanes in a limited number of candidate solvents.

44.  $\text{Si}_{5+}$  cyclosilane printability data were collected for a variety of solvents, including cycloalkanes, arenes, a linear alkane, cyclic siloxanes, ethers, and alcohols (see page 2 of Exhibit E). Boiling point, viscosity, and  $\text{Si}_{5+}$  cyclosilane solubility and stability data are reported for the solvents as known or determined. The stability of  $\text{Si}_{5+}$  cyclosilanes in a solvent was determined by Fourier transform infrared (FTIR) absorption spectroscopy, by observing the presence or absence of characteristic Si-O bond spectral absorptions in the spectrum. Some samples were heated to 100 °C or 150 °C prior to FTIR absorption spectroscopy. For three solvents (chosen at least in part for reasons relating to the solubility of the  $\text{Si}_{5+}$  cyclosilane(s) in the solvent or the spin coating properties of the composition), characteristics of a thin film formed from UV polymerization of an ink containing the  $\text{Si}_{5+}$  cyclosilane(s) in the tested solvent are reported.

45. Cycloalkanes (cyclooctane, *cis*-decalin, and mixed decalins) provide adequate boiling point, viscosity,  $\text{Si}_{5+}$  cyclosilane solubility and stability, and UV-polymerized thin film properties. Arenes generally exhibited similar viscosity,  $\text{Si}_{5+}$  cyclosilane solubility and stability

results. Cyclic siloxanes generally exhibited similar viscosity, Si<sub>5+</sub> cyclosilane solubility and stability results.

46. Ethers and alcohols generally exhibited relatively poor Si<sub>5+</sub> cyclosilane solubility and stability results, although the viscosities were generally higher (and thus, more suitable for printing applications). The unacceptable Si<sub>5+</sub> cyclosilane solubility and stability results observed for ethers and alcohols was unexpected, given that the published literature discloses mixtures of Si<sub>5+</sub> cyclosilane(s) in such ethers and alcohols.

47. Terpinen-4-ol and terpineol generally exhibited adequate Si<sub>5+</sub> cyclosilane solubility and stability results relative to cycloalkanes, arenes, and cyclic siloxanes. Terpinen-4-ol and terpineol also generally exhibited higher viscosities relative to cycloalkanes, arenes, and cyclic siloxanes. However, UV-polymerized thin films formed from solutions of Si<sub>5+</sub> cyclosilanes in terpinen-4-ol and terpineol exhibited some oxidation (i.e., Si-O bonds in the FTIR spectrum and/or oxygen detected in an atomic absorption spectrum of material from the thin films), and thus, were unacceptable for use in Si<sub>5+</sub> cyclosilane-containing inks for making thin silicon films.

48. UV polymerization trials of 5 wt% solutions of Si<sub>5+</sub> cyclosilanes in either cyclooctane or o-xylene yielded similar results for both solvents (see page 3 of Exhibit E).

49. However, cycloalkane solvents (cyclooctane and *cis*-decalin) generally solubilized viscous silanes (generally, Si<sub>5+</sub> cyclosilanes further containing some UV oligomerized and/or polymerized Si<sub>5+</sub> cyclosilanes), whereas aromatic solvents (o-xylene or methylnaphthalene) generally did not solubilize viscous silanes (see page 4 of Exhibit E).

50. To be suitable for inkjet printing, the solvent for a silane-containing ink should provide the ink with an appropriate viscosity and surface tension (see page 5 of Exhibit E). The solvent must also solubilize Si<sub>5+</sub> cyclosilanes and polymerized silanes to enable formation of silicon films having acceptable physical and electrical properties.

51. Terpinen-4-ol, other alcohol and ether solvents were determined to have unacceptable properties for cyclosilane ink printing applications (see page 5 of Exhibit E).

52. I also conducted, supervised, witnessed or otherwise know from the ordinary course of business at Kovio a number of experiments performed at Kovio to test the suitability of various solvents for inkjet printing of Si<sub>5+</sub> cyclosilane solutions.

53. A technical report presented by my colleagues and I to Kovio, Inc. management during the ordinary course of business (entitled *Solvent Compatibility Tables*; attached hereto as Exhibit F) reports the results of tests performed to determine the suitability of various solvents for inkjet printing of silane mixtures (containing approximately 80% linear and branched polysilanes and approximately 20% Si<sub>5+</sub> cyclosilanes).

54. Candidate solvents (see page 2 of Exhibit F) from five representative solvent classes (alkanes, arenes, ethers, halogenated alkanes and alkylsilanes) were purified over activated alumina, and the solubility of silane mixtures was tested for each solvent.

55. None of the tested solvents solubilized the silane mixture (see page 2 of Exhibit F). However, cycloalkane solvents (e.g., *cis*-decalin, cyclooctane and cyclodecane) were again noted to have superior performance in solubilizing the same mixtures (see page 2 of Exhibit F), and generally yielded silicon films having sufficiently good electronic properties to form electrically operational thin film transistors (TFTs), diodes, and/or other electronic structures therefrom (see page 3 of Exhibit F).

56. The data, results and observations described in paragraphs 40-41, 44-51 and 54-55 above, the disclosures of Shiho and Jacobson '401, along with other results and data with which I am familiar as a result of my experience in this area of silane chemistry, demonstrate unexpected and superior Si<sub>5+</sub> cyclosilane solubility, stability and UV polymerization properties in cycloalkanes relative to other solvents.

57. Based on my experience in silane chemistry and silicon and metal nanoparticles, the results described in paragraphs 40-41, 44-51 and 54-55 above, and discussed in this Declaration are not expected to change significantly as a result of the mere presence of passivated silicon-containing semiconductor nanoparticles in the composition containing one or more  $\text{Si}_5+$  cyclosilanes and a cycloalkane solvent.

58. I understand that a third independent claim of the above-identified application is directed to a method of making a patterned semiconductor film, comprising the steps of:

- a) printing a solution comprising passivated semiconductor nanoparticles, a first cyclic Group IVA compound of the formula (1):



where n is from 3 to 8, each of the n instances of x is independently 1 or 2, and each A in the formula is independently Si or Ge, and a solvent in a pattern on a substrate, wherein said pattern comprises one or more lines having a width of not more than 100  $\mu\text{m}$ , a length of not more than 5000  $\mu\text{m}$ , a thickness of not more than 1000  $\mu\text{m}$ , and an inter-line spacing of not more than 100  $\mu\text{m}$ ; and

- b) curing said printed pattern to form said patterned semiconductor film.

59. The mass loading of silicon-containing semiconductor nanoparticles in a liquid-phase composition comprising the silicon-containing semiconductor nanoparticles (which may also include cyclic Group IVA compounds) is critical to enable formation of patterned semiconductor films having well-defined features which display controllable, reproducible morphology. Insufficient mass loading of silicon-containing semiconductor nanoparticles may result in beading up, evaporation, or other phenomena which may result in uneven spreading of the printed ink, resulting in unacceptable film morphology and/or pattern uniformity, thickness uniformity and reproducibility.

60. Similarly, the stability of cyclic Group IVA compounds in a liquid-phase composition comprising cyclic Group IVA compounds (which may also include passivated

silicon-containing semiconductor nanoparticles) and a solvent is important in making patterned semiconductor films. If the cyclic Group IVA compounds are not stable in the composition, they may precipitate prior to printing, or during a printing process, resulting other phenomena (e.g., clogged inkjet nozzles or uneven mass distribution) which may produce undesirable variations in a patterned semiconductor film.

61. Furthermore, to be suitable for printing, the solvent for a solution comprising passivated semiconductor nanoparticles and a cyclic Group IVA compound must (i) provide the ink with an appropriate viscosity and surface tension, (ii) solubilize the Group IVA compound (e.g., a Si<sub>3</sub>+ cyclosilane), and (iii) solubilize the passivated silicon-containing semiconductor nanoparticles to enable formation of patterned semiconductor films having well-defined features (e.g., with pattern dimensions as described in paragraph 58 above) which display controllable, reproducible pattern dimensions and morphology.

62. Shiho discloses forming a silicon film on a substrate by forming a coating film of a silane composition on the substrate by means such as spray coating, roll coating, curtain coating, spin coating, screen printing, offset printing or ink jet printing (see paragraphs [0106] and [0110] of Shiho). Shiho further discloses that a silicon film, conductive film and insulating film may be formed and patterned before use by a general method such as masking or lithography, or by an inkjet method (see paragraph [0153] of Shiho). Shiho then discloses that the thickness of a silicon film is preferably 0.005 μm to 20 μm, and more preferably 0.01 to 10 μm (see paragraph [0122] of Shiho; emphasis added).

63. Shiho is silent with respect to passivated silicon-containing semiconductor nanoparticles (see paragraph 58 above). Shiho is also silent with respect to one or more lines having a width of not more than 100 μm, a length of not more than 5000 μm, a thickness of not more than 1000 μm, and an inter-line spacing of not more than 100 μm (see paragraph 58 above; emphasis added).

64. Therefore, Shiho cannot recognize that the mass loading of silicon-containing semiconductor nanoparticles in a liquid-phase composition comprising the silicon-containing semiconductor nanoparticles (which may also include cyclic Group IVA compounds) is critical to enable formation of patterned semiconductor films having well-defined features (e.g., lines having dimensions as described in paragraph 58 above) which display controllable, reproducible pattern dimensions and morphology.

65. Jacobson '401 is silent with respect to cyclic Group IVA compounds. Jacobson '401 is also silent with respect to any dimensions of a printed pattern or features, such as one or more lines having a width of not more than 100  $\mu\text{m}$ , a length of not more than 5000  $\mu\text{m}$ , a thickness of not more than 1000  $\mu\text{m}$ , and an inter-line spacing of not more than 100  $\mu\text{m}$  (see paragraph 58 above).

66. Therefore, Jacobson '401 cannot recognize that the stability of compositions containing  $\text{Si}_{5+}$  cyclosilanes are important for enabling printable compositions containing one or more  $\text{Si}_{5+}$  cyclosilanes (which may also include passivated silicon-containing semiconductor nanoparticles; see paragraph 58 above) for making patterned semiconductor films having well-defined features (e.g., lines having dimensions as described in paragraph 58 above) which display controllable, reproducible pattern dimensions and morphology.

67. The observed solubility of alkyl-passivated silicon-containing semiconductor nanoparticles in an arene solvent and an ether solvent (see paragraphs 18 and 20 above), and the observed stability of  $\text{Si}_{5+}$  cyclosilanes in some cycloalkane solvents relative to solvents such as arenes, alcohols, ketones, alkenes, ethers, and fluorinated solvents (see paragraph 40 above) are not complementary.

68. For example, alkyl-passivated silicon nanoparticles exhibit a solubility of greater than 5% in butyl ether (see paragraph 20 above). However,  $\text{Si}_{5+}$  cyclosilanes were observed to be unstable or insoluble in butyl ether (cf. page 3 of Exhibit D). Similarly, alkyl-passivated silicon nanoparticles exhibited solubility of up to 5% in xylene (see paragraph 20 above).

However, in some experiments,  $\text{Si}_{5+}$  cyclosilanes were observed to be soluble in xylene (see page 2 of Exhibit E), while in other experiments,  $\text{Si}_{5+}$  cyclosilanes were observed to produce a white precipitate in xylene then dissolution over time, suggesting that  $\text{Si}_{5+}$  cyclosilanes may not be entirely stable in xylene (cf. page 3 of Exhibit D).

69. Since Shiho is silent with respect to passivated silicon-containing semiconductor nanoparticles, Shiho cannot recognize the non-complementary results (see paragraphs 67 and 68 above) observed in efforts to develop a method of making a patterned semiconductor film including printing a solution comprising passivated semiconductor nanoparticles, a cyclic Group IVA compound and a solvent in a pattern on a substrate., where the pattern comprises one or more lines having a width of not more than 100  $\mu\text{m}$ , a length of not more than 5000  $\mu\text{m}$ , a thickness of not more than 1000  $\mu\text{m}$ , and an inter-line spacing of not more than 100  $\mu\text{m}$  (see paragraph 58 above).

70. Furthermore, since Jacobson '401 is silent with respect to a cyclic Group IVA compound, Jacobson '401 cannot recognize the non-complementary results (see paragraphs 67 and 68 above) observed in efforts to develop a method of making a patterned semiconductor film including printing a solution comprising passivated semiconductor nanoparticles, a cyclic Group IVA compound and a solvent in a pattern on a substrate, where the pattern comprises one or more lines having a width of not more than 100  $\mu\text{m}$ , a length of not more than 5000  $\mu\text{m}$ , a thickness of not more than 1000  $\mu\text{m}$ , and an inter-line spacing of not more than 100  $\mu\text{m}$  (see paragraph 58 above).

71. The non-complementary solubility results for alkyl (e.g., dodecyl) passivated silicon-containing semiconductor nanoparticles in certain solvents (see, e.g., paragraph 20 above) and stability results observed for  $\text{Si}_{5+}$  cyclosilanes in certain solvents (see, e.g., paragraph 40 above) observed in efforts to develop a method of making a patterned semiconductor film suggest that efforts to combine the disclosures of Shiho and Jacobson '401 would not be expected to afford a printable, stable composition comprising passivated silicon-containing semiconductor nanoparticles, a cyclic Group IVA compound, and a solvent for use in the present

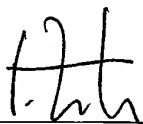


method of making a patterned semiconductor film (see paragraph 58 above), since the stability and printability of such compositions is critical in forming patterned films having well-defined features which display controllable, reproducible morphology, such as one or more lines having a width of not more than 100  $\mu\text{m}$ , a length of not more than 5000  $\mu\text{m}$ , a thickness of not more than 1000  $\mu\text{m}$ , and an inter-line spacing of not more than 100  $\mu\text{m}$  (see paragraph 58 above).

72. Accordingly, prior to the work described herein toward developing printable compositions including passivated semiconductor nanoparticles, a cyclic Group IVA compound and a solvent, it was not known whether such compositions could be formed with (i) sufficient mass loading of silicon-containing semiconductor nanoparticles, (ii) sufficient stability and solubility characteristics regarding cyclic Group IVA compounds, and (iii) suitable fluid mechanical properties for printing in a pattern having well-defined features which display controllable, reproducible morphology, such as one or more lines having a width of not more than 100  $\mu\text{m}$ , a length of not more than 5000  $\mu\text{m}$ , a thickness of not more than 1000  $\mu\text{m}$ , and an inter-line spacing of not more than 100  $\mu\text{m}$  (see paragraph 58 above).

73. Further, Declarant sayeth not.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the above-identified application or any patent issued thereon or therefrom.

  
\_\_\_\_\_  
Fabio ZÜRCHER

11/10/2009  
\_\_\_\_\_  
Date